Raman scattering in hexagonal $Al_xGa_{1-x}N$ alloys and optical modes behavior

V.Yu.Davydov[1], A.A.Klochikhin[1], I.N.Goncharuk[1], A.N.Smirnov[1], A.E.Nikolaev[1], A.S.Usikov[1], W.V.Lundin[1], M.V.Baidakova[1], J.Aderhold[2], J.Stemmer[2], and O.Semchinova[2]
[1]*Ioffe Physico-Technical Institute, St Petersburg 194021, Russia*
[2]*LFI Universitat Hannover, Schneiderberg 32, 30167 Hannover, Germany*
e-mail: Valery.Davydov@pop.ioffe.rssi.ru

We present the experimental Raman data on dynamic properties of $Al_xGa_{1-x}N$ alloys in the entire compositional range ($0<x<1$). Special attention is given to the behavior of phonon modes in Al-rich alloys. The investigation is supported by the theoretical approach describing changes in the vibration spectrum at a sufficiently strong perturbation producing a local or gap mode.

To trace the mode behavior in more detail, we used a large set of samples with the difference in the Al content not more than 3-5% in the entire compositional range. The 0.5-2 μm thick $Al_xGa_{1-x}N$ layers in the composition range $0<x<0.6$ were grown on the c-plane sapphire substrates by MBE and MOCVD techniques. The 1-2 μm thick layers of $Al_xGa_{1-x}N$ alloys in the composition range $0.6<x<1$ were grown by HVPE on a (111) silicon substrate. The structural quality of the layers and the alloy composition were controlled by X-ray diffraction and electron probe microanalysis.

There are six optical modes A_1(TO), A_1(LO), E_1(TO), E_1(LO), E_2(high), and E_2(low) active in the first-order Raman scattering of hexagonal $Al_xGa_{1-x}N$. On the whole, the measured polarized Raman spectra were found to be consistent with the selection rules for the wurtzite structure in the entire compositional range $0<x<1$. At the same time, we have found that the general pattern of the mode behavior for the Al-rich compositional range is much more complicated.

Our findings clearly confirm the theoretical predictions [1,2] that both longitudinal modes must exhibit a one-mode type behavior. We have found that the frequency positions of the A_1(LO) and E_1(LO) phonons are different in the whole compositional range, which is not consistent with [3] where the same frequencies of the A_1(LO) and E_1(LO) phonons for $0.5<x<0.7$ were given.

Our experiments confirmed the two-mode behavior of the nonpolar E_2(high) phonon observed in [3,4]. In addition, we could trace in more detail the behavior of the E_2 (high) phonon in the range of very high Al contents. At extremely high Al concentrations ($x=0.98$), narrowing of the GaN-like E_2(high) phonon line takes place. This points to a preferentially localized nature of this vibration. We have also obtained the data pointing to the two-mode type behavior of E_2(low) phonon mode.

A convincing evidence of the two-mode behavior of the E_1(TO) mode has been obtained, consistent with recent theoretical predictions [2] . Our data agree well with the IR reflection data [5] for the GaN-like E_1(TO) mode and considerably differ from the IR data for the AlN-like E_1(TO) mode.

The Raman spectrum of A_1(TO) mode for $Al_xGa_{1-x}N$ with a Ga content $(1-x)<0.3$ has been found to exhibit a large broadening with a complex structure. We attribute this structure to manifestation of the phonon density of states in the region of vibrations of the optical A_1(TO) branch and appearance of the gap mode in AlN. Both effects are due to the substitution of heavier Ga atoms in the cation sublattice of AlN. We suggest the theoretical approach describing changes in the vibration spectrum at a sufficiently strong perturbation resulting from the isoelectron substitution. In the framework of the developed model, the dependence of the intensity and band shape of the gap mode on Ga content was calculated and compared with the experimental Raman data. The results indicate that the formation of gap A_1(TO) mode in the regions of low and intermediate Ga contents is caused by statistical Ga clusters in the cation sublattice of the alloy. In a limited range of Ga contents the behavior of the A_1(TO) phonon mode in $Al_xGa_{1-x}N$ can be considered as two-mode one.

To summarize, a comprehensive study of the behavior of all six optical phonon modes in Raman spectra of hexagonal $Al_xGa_{1-x}N$ alloys has been carried out. The two-mode type behavior of the nonpolar E_2(high) phonon and one-mode type behavior of the A_1(LO) and E_1(LO) phonons has been confirmed, and their behavior for Al-rich compositions has been traced in detail. For the first time, the two-mode behavior of TO phonons with A_1 and E_1 symmetry, as well as nonpolar E_2(low) phonon has been revealed in Raman spectra. Changes in the Raman spectrum have been described using the theoretical model based on the microscopical approach. The experimental and theoretical dependences have been shown to be in good agreement.

This work is supported by RFBR (Grant No.99-02-18318) and the Program "Physics of Solid State Nanostructures".

References

[1] S.G.Yu, K.W.Kim, L.Bergman, M.Dutta, M.A.Stroscio, and J.M.Zavada, Phys.Rev. **B58**, 15283 (1998).
[2] H.Grille, Ch.Schnittler, and F.Bechstedt, Phys.Rev. **B61**, 6091 (2000).
[3] F.Demangeot, J.Groenen, J.Frandon, M.A.Renucci, O.Briot, S.Clur, and R.L.Aulombard, Appl.Phys.Lett. **72**, 2674 (1998).
[4] A.Cros, H.Angerer, R.Handschuh, O.Ambacher, and M.Stutzmann, Solid State Commun. **104**, 35 (1997).
[5] P.Wisniewski, W.Knap, J.P.Malzac, J.Camassel, M.D.Bremser, R.F.Davis, and T.Suski, Appl.Phys.Lett. **73**, 1760 (1998).